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Published in:
Europhysics Letters

DOI:
[10.1209/epl/i1999-00533-6](https://doi.org/10.1209/epl/i1999-00533-6)

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Document Version
Publisher's PDF, also known as Version of record

Publication date:
1999

[Link to publication in University of Groningen/UMCG research database](#)

Citation for published version (APA):

Sherman, E. Y., Fischer, M., Lemmens, P., Loosdrecht, P. H. M. V., & Güntherodt, G. (1999). Electron-phonon and spin-phonon coupling in NaV₂O₅: Charge fluctuations effects. *Europhysics Letters*, 48(6), 648 - 654. <https://doi.org/10.1209/epl/i1999-00533-6>

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Electron-phonon and spin-phonon coupling in NaV_2O_5 : Charge fluctuations effects

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(received 16 March 1999; accepted in final form 15 October 1999)

PACS. 63.20Kr – Phonon-electron and phonon-phonon interactions.

PACS. 75.30Et – Exchange and superexchange interactions.

PACS. 78.30–j – Infrared and Raman spectra.

Abstract. – We show that the asymmetric crystal environment of the V site in the ladder compound NaV_2O_5 leads to a strong coupling of vanadium $3d$ electrons to phonons. This coupling causes fluctuations of the charge on the V ions, and favors a transition to a charge-ordered state at low temperatures. In the low temperature phase the charge fluctuations modulate the spin-spin superexchange interaction, resulting in a strong spin-phonon coupling.

Introduction. – The spin-Peierls transition in one-dimensional spin systems is one of the best known and investigated cooperative phenomena in low-dimensional physics. This transition comprises a dimerization of the lattice of a Heisenberg spin chain, leading to an alternation of the exchange parameter J . As a result, nearest-neighbor spins form a singlet state accompanied by the opening of a spin gap. Consequently, the static spin susceptibility drops drastically below the transition temperature (T_{SP}). This effect, predicted by Pytte [1], was first observed in one-dimensional organic compounds [2]. The discovery was followed by theoretical work of Cross and Fisher [3], who showed a possibility to calculate T_{SP} from the coupling of spins to the lattice. The discovery of the spin-Peierls transition in the inorganic spin-chain compound CuGeO_3 [4] renewed interest in this phenomenon. In this half-filled chain compound each Cu site holds one “hole” with spin $s = 1/2$. The spin-spin exchange interaction occurs via oxygen orbitals. Lattice vibrations cause a modulation of the spin-spin superexchange due to changes in the inter-ionic distances and bond angles. This leads to a spin-lattice coupling which plays an important role as a driving force of the spin-Peierls transition.

The ladder system NaV_2O_5 was proposed to be a second inorganic spin-Peierls compound [5]. It contains VO_5 pyramids which form V-O ladders extended along the b -axis [6], see fig. 1a). For half the ladders the apical oxygen O3 is above the V ions while for the other half they are below them. From the chemical formula one finds an average vanadium valence of +4.5. This implies that a $3d$ electron is shared between two V ions within one rung, making NaV_2O_5 a “quarter-filled” ladder compound [7]. The shared electron introduces an additional degree of freedom: there is a possibility for the electron to move between the right

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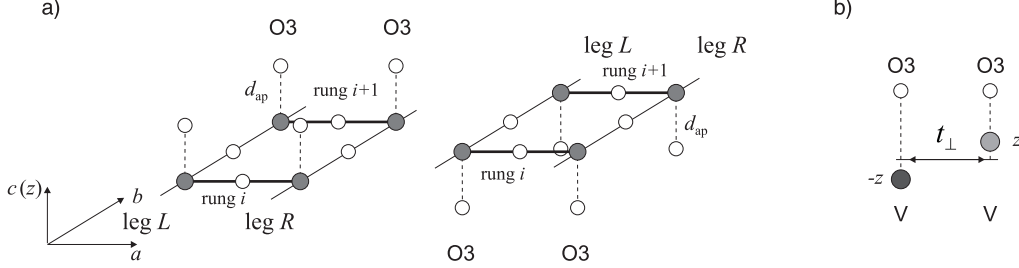


Fig. 1. – a) Schematic plot of a part the crystal structure of the NaV_2O_5 compound. Grey circles denote V ions while open circles correspond to the O ions. The V-O-V rungs are shown in bold. b) Charge disproportionation arising due to out-of-phase V ion displacements along the z -axis. The light-grey (dark-grey) circle corresponds to the decreased (increased) electronic density on the site.

(R) and left (L) V ions within one rung. This feature leads to physical differences between NaV_2O_5 and CuGeO_3 , the most striking one being a possible *charge* ordering in NaV_2O_5 . If an additional degree of freedom is involved in a phase transition, one should expect enhancement of the anomalies accompanying it. In fact, available experimental data on NaV_2O_5 reveal strong anomalies in heat conductivity [8], specific heat [9], inelastic neutron scattering [10], and inelastic light scattering [11]. These observations can be considered as i) confirmation of changes in the electronic subsystem in addition to a spin gap opening, and ii) indirect evidence for charge ordering. Direct evidence of charge separation within a rung was provided by a nuclear magnetic resonance experiment [12] which showed that two non-equivalent V ions exist below T_C . In addition, X-ray and elastic neutron scattering revealed a lattice superstructure with a modulation vector $(1/2, 1/2, 1/4)$ in the low-temperature phase [13]. However, the nature of the charge ordering and the strength of the charge asymmetry are not clear yet.

Two different models for the charge-ordered phase were introduced recently, notably an “in-line” [14] and a “zigzag” structure [15, 16]. In the in-line structure all V ions within one leg have the same charge Q_{il} but $Q_{iL} \neq Q_{iR}$, where i enumerates rungs along the ladder, while $l = L, R$ denotes the sites within one rung. For the zigzag state one has $Q_{iR} = Q_{i+1,L}$. For both the states the electron moves within one rung, *i.e.* $Q_{iL} + Q_{iR} = 1$. It was proposed in [14] that the phase transition occurs in two stages. First the charge subsystem orders, which is followed by the opening of a spin gap. Mostovoy and Khomskii [16] argued that a zigzag ordering itself may lead to the opening of a spin gap due to an induced alternation of the exchange within the ladders. Nevertheless, coupling to the lattice is always important for the spin-Peierls transition either as a driving force or as a factor stabilizing the charge ordering [16]. For a better understanding of the role the lattice plays it is important to know the values and physical origin of the parameters involved in a model taking the spin, charge and lattice degrees of freedom into account.

Here we investigate a mechanism of electron-phonon coupling in NaV_2O_5 and determine the scale of the parameters of the spin-phonon interaction. Due to the asymmetric structural environment of the V sites (see fig. 1a)) vibration-induced shifts of the electron site energies are proportional to the z -axis displacements of the ions. The small V-O3 equilibrium distance $d_{\text{ap}} \approx 1.61 \text{ \AA}$ [7] leads to a strong contribution of the O3 ion Coulomb field, E_{ap} , to the $3d$ electron site energy. A phonon mode involving a non-equivalent modulation of d_{ap} on the right and left side therefore causes a charge redistribution between the V ions on a rung. It will be shown that in the charge-ordered phase this effect leads to a strong spin-phonon

coupling activated by the charge disproportion. The Coulomb field enhances the role of electronic correlations proposed to be responsible for the charge ordering [14-16] and leads to phonon-driven fluctuations of the exchange. Finally, two possible experimental consequences of the charge disproportion and fluctuations for optical spectroscopy will be analyzed.

One-rung charge disproportion. – Consider a V-O-V rung, denoting the indirect hopping between the V ions as $t_{\perp} \approx 0.35$ eV [7, 17]. Then there are two in-rung states separated in energy by $2t_{\perp}$: an odd and an even one, and both V ions have charge $+4.5$. Now consider an out-of-phase vibration of the two V ions along the z -axis (see fig. 1b)). The corresponding zero-point amplitude is $z_0 = \sqrt{\hbar/M\Omega}$, where M is the V ion mass, and Ω is the phonon frequency. For $\Omega = 400$ cm $^{-1}$, which is an estimate justified later by our experimental data, $z_0 \approx 0.04$ Å. Under a displacement ($\pm z$) of the V ions the on-site energies will be changed by an amount $\pm Cz$, inducing a charge transfer between the V ions (C is the deformation potential). By determining the eigenstates of this asymmetric two-level system the ground-state energy ε_0 and the amount of the charge transfer are easily calculated, yielding

$$\varepsilon_0 = -\sqrt{t_{\perp}^2 + C^2 z^2}, \quad \delta Q = Q - \frac{1}{2} = \pm \frac{Cz}{2\sqrt{t_{\perp}^2 + C^2 z^2}}, \quad (1)$$

where $Q = 0$ for V^{+5} state. A point-charge Coulomb estimate of $E_{\text{ap}} = 2e/d_{\text{ap}}^2$ yields $C = eE_{\text{ap}} = 12$ eV/Å. Therefore Cz_0/t_{\perp} , the parameter characterizing the electron-phonon coupling, is of order unity. Assuming that an adiabatic approximation is valid ($\Omega \ll t_{\perp}$), and calculating the mean value with the vibrational ground-state wave function $|\phi\rangle = (\pi z_0^2)^{-1/4} e^{-z^2/2z_0^2}$, we obtain the charge fluctuations in the charge-symmetrical (high-temperature) phase:

$$\langle \phi | (Q - 1/2)^2 | \phi \rangle = \frac{1}{4} - \frac{\sqrt{\pi}\xi}{4z_0} e^{\xi^2/z_0^2} [1 - \Phi(\xi/z_0)], \quad (2)$$

where $\xi \equiv t_{\perp}/C = 0.03$ Å, and $\Phi(\xi/z_0)$ is the error function. Since $\xi \approx z_0$, we obtain $\langle \phi | (Q - 1/2)^2 | \phi \rangle \approx 0.18$, *i.e.* the fluctuations are of the order of the net average charge.

Now we can estimate the role of the displacement-induced site asymmetry in the formation of the low-temperature phase. Suppose that inter-site Coulomb interactions, leading to intersite correlations, induce a static splitting $2\Delta_{\text{cor}}$ of the on-site levels within a rung. This splitting causes a charge disproportion $\varphi = |Q_{\text{L}} - Q_{\text{R}}|$. This purely correlation-induced disproportion φ_{cor} is of the order of $\Delta_{\text{cor}}/t_{\perp}$. However, the apical oxygen field at the d -electron considerably enhances this charge redistribution. The correlation-induced charge asymmetry will cause a difference in the Coulomb forces acting on the V ions. This difference induces static opposite displacements of the ions, leading to a further increase of the charge disproportion. Eventually, this process will stop due to the elastic energy of the lattice. The additional splitting $2\Delta_{\text{lat}}$ of the on-site levels leads to a total site energy shift $\pm\Delta = \Delta_{\text{lat}} + \Delta_{\text{cor}}$, the average ion charges are given by $Q_{\text{L,R}} - 1/2 = \pm\Delta/2 (t_{\perp}^2 + \Delta^2)^{1/2}$. The new equilibrium positions \bar{z} of the V ions are determined by a self-consistency equation:

$$x = \frac{C^2}{2t_{\perp}\tilde{k}} \cdot \frac{1+x}{\sqrt{1+\tau^2(1+x)^2}}, \quad (3)$$

where $\tau \equiv \Delta_{\text{cor}}/t_{\perp}$, $\Delta_{\text{lat}} \equiv C\bar{z}$, $x \equiv \Delta_{\text{lat}}/\Delta_{\text{cor}}$, and \tilde{k} the non- d -electron contribution to the total force constant $k = M\Omega^2$ (≈ 30 eV/Å 2) relevant for the vibration. Equation (3) is a consequence of the equilibrium condition $\tilde{k}\bar{z} = C\varphi/2$ and eq. (1) for the charge transfer

between the V ions. It has the following limiting cases for $\tau \ll 1$:

$$x = \begin{cases} A/(1-A), & 1-A^2 \gg \tau^2, \\ 2^{1/3}\tau^{-2/3}-1, & 1-A^2 \ll \tau^2, \end{cases} \quad (4)$$

with $A \equiv C^2/2t_\perp \tilde{k}$. The first limit corresponds to a response of the lattice to a relatively small correlation-induced charge disproportion. The second one shows that if the lattice is close to an instability ($A \approx 1$) even weak correlations induce a large charge disproportion. The d -electron contribution to the force constant can be calculated from eq. (1) as $k_d = \partial^2 \varepsilon_0 / 2\partial z^2 \approx -C^2/2t_\perp$ (≈ -200 eV/Å² for $C = eE_{\text{ap}}$), yielding $A = -k_d/(k - k_d) \approx 0.85$. It follows from eq. (4) that x can be larger than unity, in which case the main contribution to φ comes from the apical oxygen (O3) field. This field enhances the correlation-induced asymmetry of the V ions and thus favors charge ordering.

For $\tau > 1$ eq. (3) yields $x \sim 1/\tau$. The charge disproportion and ion displacement now saturate at $\varphi \approx 1$ and $\bar{z} \sim C/\tilde{k}$, respectively. The corresponding \bar{z} is of the order of z_0 , in agreement with the experimental shifts reported in ref. [18]. The corresponding site energy shift is of the order of $\bar{z}C \sim t_\perp$. Within a linear approximation the phonon-induced charge transfer in the ordered phase can be written as

$$\delta Q = C\delta z \frac{t_\perp^2}{(t_\perp^2 + \Delta^2)^{3/2}}, \quad (5)$$

where δz is the out-of-phase displacement from the new equilibrium positions. As we will see below, these charge fluctuations lead to a spin-phonon coupling in the low-temperature phase.

Phonon-induced fluctuations of the exchange. – It has been proposed that above T_c the spin system of NaV₂O₅ can be mapped onto an antiferromagnetic Heisenberg chain with $\hat{H} = J \sum \mathbf{s}_i \mathbf{s}_{i+1}$, where \mathbf{s}_i is the spin *per rung* [7, 17] and $J \approx 560$ K [5]. Using this idea, we can calculate J within a simple model. The superexchange occurs mainly due to hopping via the oxygen orbitals within the legs. We denote the hopping integral between V and O orbitals along the ladder as t_{pd} , and the one-electron energy difference for the in-leg O($2p$) \rightarrow V($3d$) transition as $\varepsilon \approx 6.5$ eV [17]. Allowing for inequivalent nearest-neighbor rungs i and $i+1$, J can be expressed as

$$J = 2 \sum_{l=L,R} \frac{t_{i,l}^2 t_{i+1,l}^2}{\varepsilon^2 t_\perp}, \quad (6)$$

where the $t_{i,l}$ and $t_{i+1,l}$ are hoppings of a hole from the V sites to the O ion between them. The values of $t_{i,l}^2$ being proportional to the probability to find the hole at a corresponding V site can be readily expressed via the charges of the ions: $t_{i,l}^2 = t_{pd}^2 Q_{i,l}$. From eq. (6) we obtain $J = 2t_{pd}^4 (Q_{i,L}Q_{i+1,L} + Q_{i,R}Q_{i+1,R})/\varepsilon^2 t_\perp$ ⁽¹⁾. The charge-ordering modified exchange \tilde{J} for the in-line and zigzag phases is uniform and may be written as

$$\tilde{J} = \frac{J}{t_\perp^2 + \Delta^2} \cdot \begin{cases} t_\perp^2, & \text{zigzag phase, } Q_{i,L} = Q_{i+1,R}, \\ 2\Delta^2 + t_\perp^2, & \text{in-line phase, } Q_{i,L} = Q_{i+1,L}. \end{cases} \quad (7)$$

⁽¹⁾ Expressing J as a function of the charges is a common feature of the approaches based on perturbation theory [19].

The charge separation in the zigzag phase decreases the exchange while the in-line ordering increases it [19]. These effects can be easily understood since in the zigzag phase the electrons are more separated in space while in the in-line phase they are closer to each other. The experimentally found $\tilde{J}/J \approx 0.8$ [20] thus is indicative of a zigzag type of charge ordering in NaV_2O_5 .

The phonon causing charge fluctuations also modulates the hopping $t_{i,l}$ and, in turn, the exchange \tilde{J} . For phonon momentum q along the ladder the modulation of \tilde{J} is found to be

$$\frac{\delta\tilde{J}}{\delta z} = 4J(Q_L - Q_R)f(q)\frac{\delta Q}{\delta z}, \quad (8)$$

with $f(q) = i\sin(qd/2)$ and $f(q) = \cos(qd/2)$ for the zigzag and in-line phases, respectively. δQ is the charge transfer in eq. (5) and d the in-ladder lattice constant. This contribution to the spin-phonon coupling is solely due to charge asymmetry and therefore a characteristic feature of the low-temperature phase. Following the approach of Pytte [1], we estimate the spin-phonon coupling constant as $\lambda \sim z_0^2(\delta\tilde{J}/\delta z)^2(\Omega\tilde{J})^{-1}$. Note that, within the present model, a complete charge ordering ($\varphi = 1$, $\Delta \gg t_\perp$) suppresses spin-phonon coupling since it prohibits phonon-induced charge fluctuations (see eq. (5)). From eqs. (5)-(8) we obtain

$$\lambda \sim \frac{C^2 z_0^2 J}{t_\perp^2} \frac{\Delta^2}{\Omega (\Delta^2 + t_\perp^2)^3}. \quad (9)$$

The maximal λ is reached at $\Delta = t_\perp/\sqrt{2}$, corresponding to $\varphi = 1/\sqrt{3}$, $\tilde{J}/J = 2/3$, and $\lambda \sim 1$.

Finally, we address the influence of charge ordering on possible frustrating next-nearest-neighbor interactions (J'). This is interesting since frustration (J'/J) may be a driving force for the opening of a spin gap. If we suppose that both nearest and next-nearest couplings are determined by the charge order, we obtain for the zigzag phase: $\tilde{J} = J(1 - \varphi^2)$ [19] and $\tilde{J}' = J'[(1 - \varphi)^2 + (1 + \varphi)^2]/2 = J'(1 + \varphi^2)$. Zigzag ordering apparently leads to an enhancement of the role of frustration.

Experimental consequences. – The effects of the phonon-induced charge fluctuations and charge disproportion directly influence the electronic and lattice properties of the crystal and should therefore manifest themselves in Raman and infrared spectroscopy.

One experimental consequence of the charge disproportion considered above is that the out-of-phase vibration of the V ions should become Raman active below T_c . The normal vectors of even (in-phase) and odd vibrations of the V ions (see fig. 1b) can be written as $\mathbf{u}_g = (1, 1)/\sqrt{2}$ and $\mathbf{u}_u = (1, -1)/\sqrt{2}$, respectively. If $\varphi \neq 0$, these vibrations are mixed and their eigenvectors can be written in the form

$$\mathbf{u}_1 = \mathbf{u}_g \cos \theta + \mathbf{u}_u \sin \theta, \quad \mathbf{u}_2 = -\mathbf{u}_g \sin \theta + \mathbf{u}_u \cos \theta. \quad (10)$$

The angle θ is determined by the charge disproportion within a rung ($\theta = 0$ at $T > T_c$). There are two reasons for Raman activity of the second vibration in eq. (10). The first one is the presence of the Raman active \mathbf{u}_g component in the \mathbf{u}_2 mode. The second reason is that charge asymmetry leads to electronically different V ions ($\text{V}^{+4.5+\varphi/2}$ and $\text{V}^{+4.5-\varphi/2}$). Therefore, displacements of these ions give different contributions to the Raman polarizability, leading to Raman activity of the \mathbf{u}_u mode. These two effects, which cannot be separated presently, lead to an intensity I_2 of the new mode proportional to $\sim \varphi^2$.

Experimental Raman data for $T = 100$ K and $T = 5$ K are presented in fig. 2. At $T = 100$ K, only a single Lorentzian mode at 420 cm^{-1} is observed. Below the phase transition, at $T = 5$ K, this mode splits into two components centered at 394 cm^{-1} and 429 cm^{-1} , respectively. The

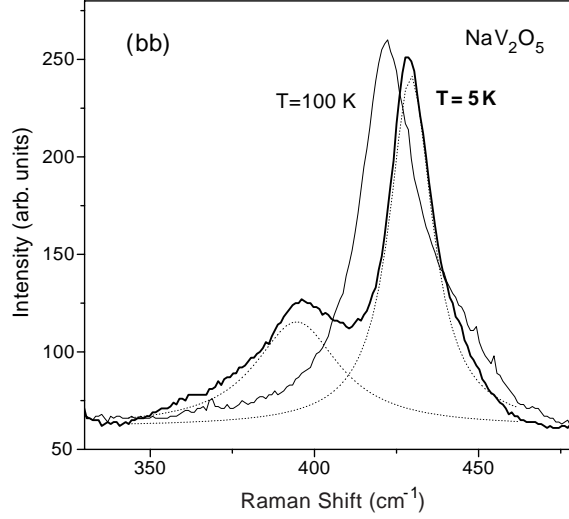


Fig. 2. – Raman spectra of NaV_2O_5 at $T = 100$ K (thin line) and $T = 5$ K (bold line) demonstrating the appearance of a new intensive phonon at 394 cm^{-1} in the low-temperature phase. Polarizations of the incident and scattered light are parallel to the ladders. The dashed line shows Lorentzian fits to the $T = 5$ K spectrum.

lower energy peak appearing for $T < T_c$ is attributed to the \mathbf{u}_2 vibration in eq. (10). The ratio of the integrated intensities of the “new” (394 cm^{-1}) and “old” (429 cm^{-1}) phonons is $I_2/I_1 \approx 0.4$. This is considerably larger than for the phonons arising solely due the superstructure formation, where the ratio is found to be smaller than 0.1 [21]. In principle, the ratio I_2/I_1 could be used to estimate the extent of the charge ordering ($\varphi \sim \sqrt{I_2/I_1}$). However, the reliability of this estimate would be questionable since the Raman polarizability of the \mathbf{u}_u is not only determined by charge ordering but is also influenced by resonance enhancement effects [22].

Another experimental consequence is related to charge fluctuations as observed in infrared (IR) experiments. IR absorption spectra at $T = 300$ K show a peak at $\hbar\omega_{\text{max}} \approx 1 \text{ eV}$ [23]. This peak has been assigned to in-rung electronic transitions. The in-rung hopping was estimated there as $t_{\perp} \approx 0.3 \text{ eV}$, whereas the difference $\hbar\omega_{\text{max}} - 2t_{\perp} \approx 0.4 \text{ eV}$ was interpreted as a result of an in-rung charge separation already at $T > T_c$. Here, we propose that this shift may be due to the electron-phonon coupling considered above, even *without* static charge separation. In the presence of the out-of-phase phonon mode the splitting of the one-rung levels is $2\sqrt{t_{\perp}^2 + C^2 z^2} > 2t_{\perp}$. Taking into account that the probability of a z -axis displacement is determined by the vibrational wave function $|\phi\rangle$, we obtain a band in the absorption spectra which starts at $2t_{\perp}$ and peaks at $\hbar\omega_{\text{max}} \approx 2t_{\perp} + \langle\phi|C^2 z^2/t_{\perp}|\phi\rangle$. Taking the same set of parameters as before, we get $\hbar\omega_{\text{max}} - 2t_{\perp} \approx C^2 z_0^2/t_{\perp} \approx 0.6 \text{ eV}$, in good agreement with the experiment. Based on this model we expect an increase of $\hbar\omega_{\text{max}}$ in the low-temperature phase, as well as a narrowing of the peak due to the suppression of charge fluctuations by the charge ordering.

Conclusion. – We investigated electron- and spin-phonon coupling in NaV_2O_5 caused by the structural vanadium site asymmetry. The electron-phonon coupling leads to charge fluctuations, and considerably increases a correlation-induced charge disproportion. The fluctuations induce a *strong* spin-phonon coupling in the charge-ordered phase. We stress,

however, that this interaction is *not* the driving force of the transition. The charge ordering at low temperatures is manifested in Raman scattering by a new intense phonon peak. The phonon-induced charge fluctuations in the high-temperature phase may explain the absorption peak observed near 1 eV in IR experiments.

Valuable discussions with C. PINETTES and M. GROVE are gratefully acknowledged. This work is supported by the DFG through SFB341, INTAS grant 96-410, and BMBF project 13N6586/8. EYaS acknowledges support from the AvH Foundation.

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